Electronic Supplementary Information for: Short range ballistic motion in fluid lipid bilayers studied by quasi-elastic neutron scattering

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1. Potential continuous diffusion at high Q-values

In a first attempt, all Q-values were fit using the continuous diffusion model assuming Lorentzian shaped quasi-elastic broadening only. The fitting procedure is described in detail in the main paper. The fitted FWHM for all Q values are shown in ***Figure ESI-1***.

This model provides a good fit for Q-values smaller than 2.65 Å^{-1}. A typical fit is shown in Figure 2b) of the main paper for a Q value of 0.52 Å^{-1}. When plotted as function of Q^2, the data points for Q values smaller than 2.65 Å^{-1} follow a linear trend, as predicted for continuous diffusion (see ***Equation 6***). The FWHM decreases and starts to deviate from the Q^2 curve for Q-values larger than 2.65 Å^{-1} (corresponding to a length scale of 2.37 Å). As the signal intensity decreases with increasing Q, the uncertainties in the high Q fits become larger, as can be seen by the error bars in ***Figure ESI-1***. The fitted values for the FWHM are consistently smaller than for the low Q and scatter around a value of 25 µeV.

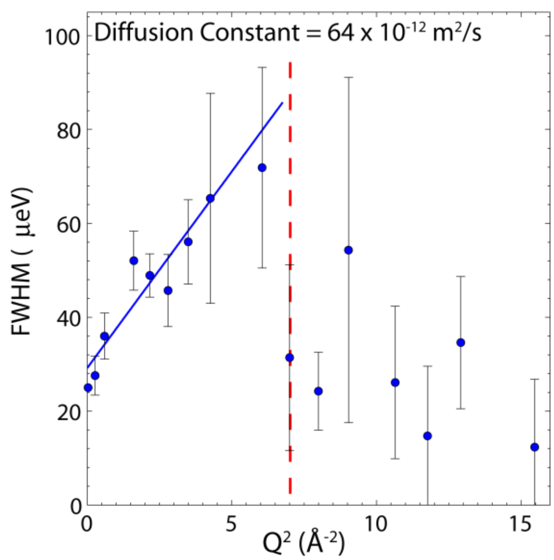

Figure ESI-1: *FWHM of Lorentzian functions used to fit all of the Q values. There is a distinct change in character of the motion occurring at ~2.37Å*

From this, two conclusions can be drawn. Firstly, the character of the motion changes at Q=2.65 Å^{-1} and the FWHM no longer follow the Q^2 behaviour predicted by the continuous diffusion model. Secondly, the high Q data in ***Figure ESI-1*** do not fall on a straight line. A second, continuous diffusion regime corresponding to the high Q data, within the dynamic window of IN13, can be excluded. Given the values of the FWHM for the high Q values in Figure ESI-1, the observed motion at high Q values is significantly slower than the continuous diffusion at small Q values.

2. *Ballistic diffusion at small distances*

Because the continuous diffusion model did not provide a satisfying description of the high Q data, a ballistic diffusion model, according to ***Equation 10***, was taken into consideration. Q values higher than 2.65 $Å^{-1}$ were fit assuming Gaussian quasi-elastic broadening. The FWHM, as determined from the fits, are shown in ***Figure 4*** in the main paper.

The ballistic model provides good quality fits with lower uncertainties as compared to the continuous diffusion model as will be discussed in the next section. The FWHM of the peaks are significantly smaller for high Q, as compared to the trend from the low Q values. We note that the observation of a critical Q-value of 2.65 $Å^{-1}$ is robust and independent of the applied model. In addition, the fitted FWHM show a linear Q dependence, as predicted by the ballistic diffusion model. Because the data at Q=2.65 $Å^{-1}$ is right at the transition between the two regimes, it was found to not follow the trend of either model; however it was better fit with a Gaussian function.

3. *Quantitative comparison between the two models*

In order to quantitatively compare the continuous and ballistic diffusion model the quality of the fits of the high Q data, using Gaussian and Lorentzian peak shapes, must be compared.

Table ESI-1 lists the uncertainties in the determination of the FWHM and χ^2 values for the Gaussian and Lorentzian fits. Due to the intrinsically large uncertainty in the data, both models can be used to fit the data. However, the Gaussian model provides mostly smaller values for the uncertainty and χ^2.

Q	χ^2 of Gaussian fit	Gaussian FWHM uncertainty (μeV)	χ^2 of Lorentzian fit	Lorentzian FWHM uncertainty (μeV)
2.6455	0.99726	10.19	1.0370	19.79
2.8278	1.1984	3.62	1.2618	8.31
3.0062	0.96619	7.20	0.97619	36.78
3.2627	1.3364	8.84	1.2795	16.25
3.4305	1.3750	20.19	1.3309	14.88
3.5935	1.0343	5.68	1.1496	14.08
3.9329	1.1858	14.51	1.1890	14.45

Table ESI-1: *χ^2 values and uncertainties (corresponding to a 95% confidence interval) in the FWHM as determined from the Gaussian and Lorentzian fits.*

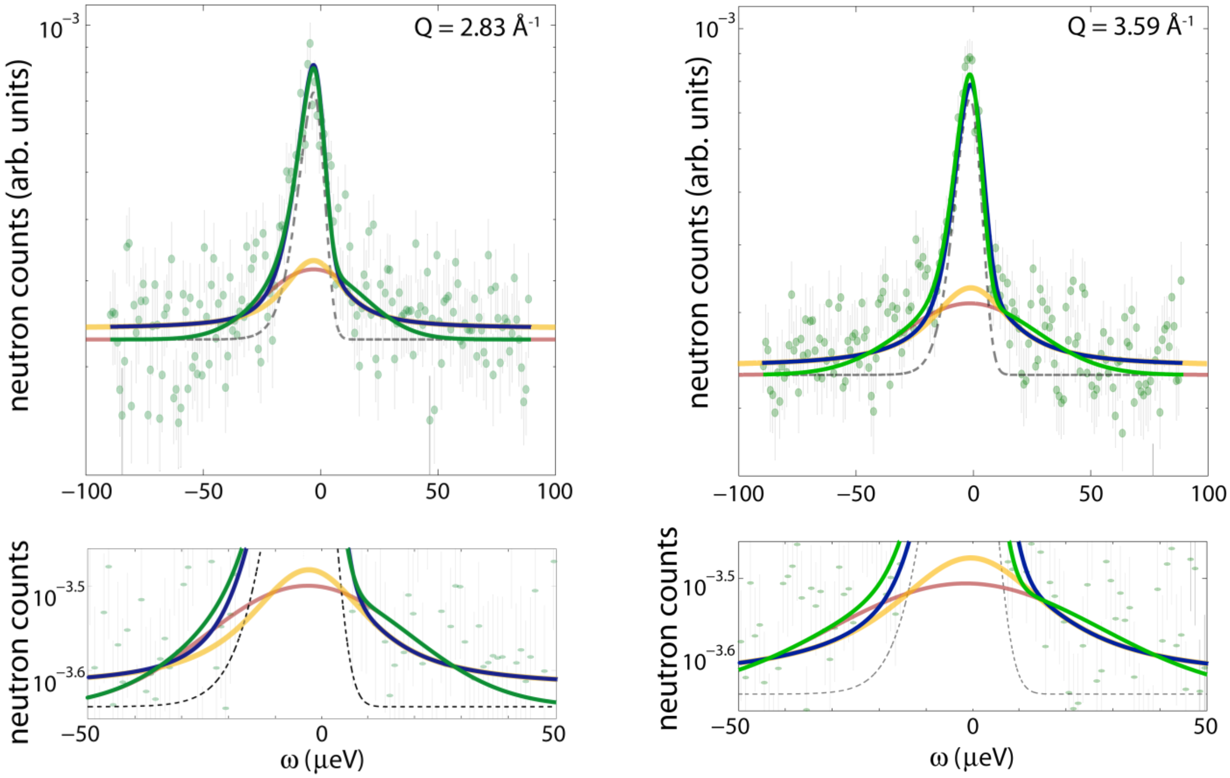

Figure ESI-2: *Comparison of the Gaussian and Lorentzian function fits at a) Q = 2.83 Å^{-1} and b) Q = 3.59 Å^{-1}. The red curve is the Gaussian function and the total Gaussian fit, with the resolution included, is the green curve. The yellow curve is the Lorentzian function, with the total Lorentzian fit shown in blue.*

Fits of two exemplary Q values are shown in ***Figure ESI-2.*** By visual inspection the Gaussian peak seems to better describe the shape of the quasi-elastic broadening at energy transfers of 10-40 μeV as the Lorentzian peak shape is narrower in the centre and falls of more quickly thereby missing the data points which the Gaussian is able to accommodate.

In summary, the dynamics at high-Q values can be fit using both a Gaussian and Lorentzian quasi-elastic broadening. In both models the fitted FWHM starts to deviate from the low-Q trend towards significantly smaller FWHM values beyond a critical Q-value of 2.65 Å^{-1}. Within the uncertainties of this experiment, the Gaussian fits were found to provide a better description of the high-Q data. The uncertainties in the fits are mostly smaller with the Gaussian function and the data show a linear Q-behaviour.

The fitted values of the FWHM scatter around 25 μeV when using the Lorentzian fits without a clear trend, such as a Q^2 behaviour. We, therefore, conclude that the ballistic motion is the appropriate model to describe the high Q data within the uncertainty of this experiment.